

IN THE UNITED STATES PATENT AND TRADEMARK OFFICE

In re Application of: SASAKI et al.) Group Art Unit: 1781
)
Application No. 10/571,266)
) Examiner: JYOTI CHAWLA
Filed: September 3, 2006)
)
For: PROCESS FOR PRODUCING) Confirmation No.: 6102
MODIFIED GUM ARABIC)
)
)
)

Commissioner for Patents
PO Box 1450
Alexandria, VA 22313-1450

Sir:

DECLARATION UNDER 37 C.F.R. § 1.132

I, Tsuyoshi KATAYAMA , do hereby make the following declaration:

1. I am a Japanese citizen, residing at 1-1-11, Sanwa-cho, Toyonaka, Osaka, 561-8588, Japan.
2. I graduated from Okayama University, Faculty of Agriculture, Department of agricultural chemistry in 1992. I also graduated from the Graduate School of Natural Science and Technology, and received a Master's Degree in 1994.
3. I also graduated from the Graduate School of Natural Science and Technology, and received a PhD Degree in 2006.
4. I began my employment with SAN-EI GEN F. F. I., INC., the assignee of the above-identified application, on April in 1994. Since 1994, I have been engaged in the research and development of Emulsion and it's emulsifier. I am in charge of research and development regarding natural emulsifier such as gum arabic.

5. I am one of the named inventors of the above-identified application, and am familiar with the subject matter of said application as well as the disclosures in the cited references.

In order to compare the present invention and the prior art teachings, I have conducted the following experiments.

Experiments

Purposes:

The purpose of this experiment is to evaluate emulsifying ability and discoloration degree of modified gum arabic samples, which are obtained by heating gum arabic samples with different particle diameters (average particle diameter: 30mm, 6mm, 1.5mm, and 0.083mm) under reduced pressure.

Methods:

(1) Materials

Four types of gum arabic (unmodified) samples with different particle diameters (samples described below) (14.5% to 14.8% of loss-on-drying) were prepared from the same gum arabic beads (*Acacia senegal*), and 300 g of each of the gum arabic samples was heated at 125°C for 3 to 12 hours while slowly rotating with a rotary evaporator at reduced pressure (about 0.03 atm. (about 3040 Pa)). For the obtained gum arabic samples, loss-on-drying (%), a discoloration degree, an emulsifying ability [average particle diameter (median diameter) (μm)], the content of arabinogalactan protein (AGP content (%)), flavor,

and appearance were measured in the same manner as in Experimental Example 1. Weight recovery ratio (%) and polydispersity (P) were determined according to the method below.

Samples: Gum arabic (unmodified)

- 1) Gum arabic beads, unpulverizates (particle diameter of 2 mm to 100 mm, average particle diameter of 30 mm)
- 2) Coarse pulverizates of gum arabic beads (particle diameter of 0.5 mm to 15 mm, average particle diameter of 6 mm)
- 3) Pulverizates of gum arabic beads (particle diameter of 0.1 mm to 2 mm, average particle diameter of 1.5 mm)
- 4) Fine pulverizates of gum arabic beads (particle diameter of 0.038 mm to 0.5 mm, average particle diameter of 0.083 mm (83 μ m))

Weight recovery ratio (%) and polydispersity (P)

Weight recovery ratio and polydispersity (P) of the gum arabic are determined by subjecting the gum arabic to be measured to GPC-MALLS under the same conditions as in Experimental Example 1, and processing the obtained data by ASTRA Version 4.5 (Wyatt Technology) software.

<Weight recovery ratio>

When all of the peaks on the chromatogram (RI chart) obtained with the RI detector under the above-described GPC-MALLS conditions were data-processed as one peak, "weight recovery ratio" used in the present invention is defined as the recovery ratio of the

one peak calculated based on weight. The aforementioned one peak on the chromatogram means the area from a starting point to an ending point, when the point where the RI chart begins to rise from the baseline of the chromatogram is defined as the "starting point", and the point where the RI chart falls and intersects the baseline is defined as the "ending point". It is known that the weight recovery ratio of one peak of unmodified gum arabic and modified gum arabic are approximately 100%. However, it is also known that since a high molecular weight component that is excessively modified and thus does not dissolve in water (which may be referred to as "hydrogel") is filtrated as insoluble matter with a 0.45- μ m membrane filter during the process for preparing the sample for GPC measurement, the weight recovery ratio of one peak falls and the emulsifying ability also lowers due to production of such high molecular weight hydrogel that is insoluble in water.

As described above, when gum arabic (unmodified) is modified, the emulsifying ability is usually improved with an increase in the above-mentioned weight average molecular weight or AGP content. However, when gum arabic (unmodified) is excessively modified, the emulsifying ability is lowered. Thus, the weight recovery ratio is used as an index for hydrogel generation.

<Polydispersity (P)>

When all of the peaks on the chromatogram (RI chart) obtained by GPC-MALLS under the above-described conditions were data-processed as one peak, polydispersity (P) is defined as the ratio of weight (converted) average molecular weight to the number (converted) average molecular weight, and thus is obtained by dividing the weight (converted) average molecular weight by the number (converted) average molecular weight. The obtained polydispersity (P) is used as a standard by which uniformity of the molecular

weight distribution is shown. The number (converted) average molecular weight and the weight (converted) average molecular weight are determined by processing the GPC-MALLS data by using ASTRA Version 4.5 (Wyatt Technology) software. The higher the P value is, the less uniform the molecular weight distribution is, while the lower the P value is, the more uniform the molecular weight distribution is. When the P value is high, the degree of modification and efficiency of the gum arabic are not satisfactory, which is shown by a nonuniformity molecular weight distribution, generation of hydrogels, lowered emulsification properties, and the like. In contrast, when the P value is low, it is considered that gum arabic is efficiently modified, which is shown by uniform molecular weight distribution, no hydrogel generation, and clearly improved emulsification properties.

(2) Experiment Results

The results are shown in Table I.

<Table I>

	Processing conditions	Loss-on-drying (%)	AGP (%)	Weight recovery ratio (%)	Polydispersity (P)	Emulsifying ability Median (μ)	Discoloration	Conditions
Samples 1) Gum arabic (unpulverizates) 30 mm	Unmodified	14.8	14.3	100	2.1	1.75	6.1	No change in appearance
	125°C, 3H at reduced pressure	11.1	20.9	96	5.2	1.23	17.1	Dried surface and wet interior
	125°C, 6H at reduced pressure	7.0	24.1	93	6.2	1.61	34.8	Dried surface and wet interior
	125°C, 12H at reduced pressure	2.7	25.0	80	6.7	3.15	36.6	Dried surface and wet interior
Samples 2) Gum arabic (coarse pulverizates) 6 mm	Unmodified	14.8	15.6	100	2.1	1.55	5.9	No change in appearance
	125°C, 3H at reduced pressure	6.5	19.3	98	4.5	1.09	25.9	Dried surface and wet interior
	125°C, 6H at reduced pressure	2.8	24.5	96	5.5	2.15	36.5	Dried surface and wet interior
	125°C, 12H at reduced pressure	1.3	26.9	93	6.0	2.84	37.5	Dried surface and wet interior
Samples 3) Gum arabic (pulverizates)	Unmodified	14.5	14.7	100	2.1	1.42	7.0	No change in appearance
	125°C, 3H at reduced pressure	2.5	18.2	99	3.8	0.81	12.1	No significant change in appearance

1.5 mm	125°C, 6H at reduced pressure	1.4	20.9	98	4.5	1.04	21.1	No change in appearance
	125°C, 12H at reduced pressure	0.3	26.5	95	5.3	1.17	25.6	No change in appearance
Samples 4) Gum arabic (Fine pulverizates) 0.083 mm	Unmodified	14.5	14.7	100	2.1	1.42	7.0	No change in appearance
	125°C, 3H at reduced pressure	0.5	18.2	100	3.6	0.79	10.5	No change in appearance
	125°C, 6H at reduced pressure	0	23.8	99	4.3	0.76	14.6	No change in appearance
	125°C, 12H at reduced pressure	0	26.6	97	5.0	1.02	19.5	No change in appearance

As is clear from Fig. A, gum arabic with a smaller particle diameter (especially, pulverizates, fine pulverizates) exhibited significantly suppressed discoloration.

Fig. B shows the relationship between the particle diameter (mm) and the emulsifying ability of gum arabic modified by heating at reduced pressure. As is clear from Fig. B, gum arabic (unmodified) to be heated having a smaller particle diameter was favorably modified (emulsifying ability is increased) while discoloration was significantly suppressed.

In contrast, it was confirmed that when gum arabic (unmodified) to be heated has a large particle diameter, discoloration is likely to proceed and such gum arabic is not uniformly modified (emulsifying ability was not improved).

Fig.A

Relationship between discoloration degree and particle diameter under heat at 125°C at reduced pressure

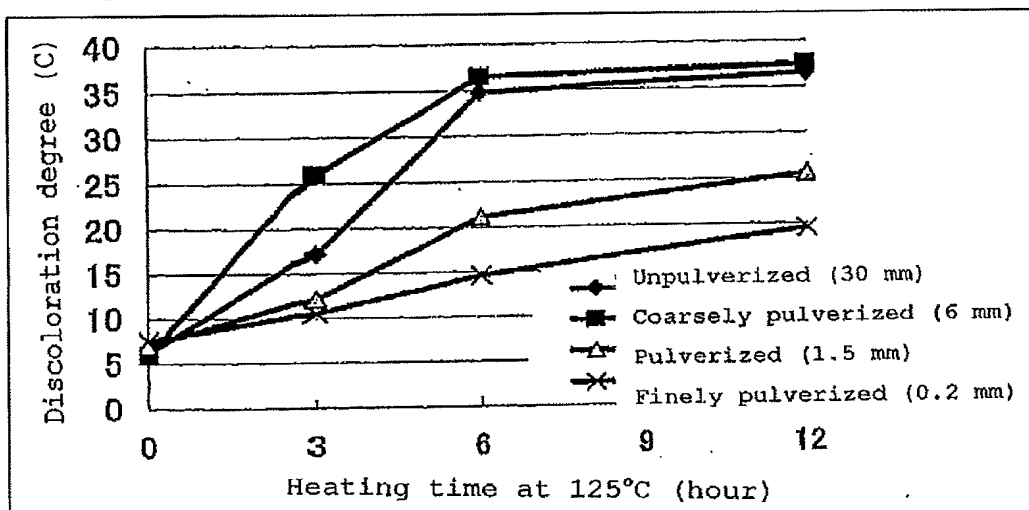
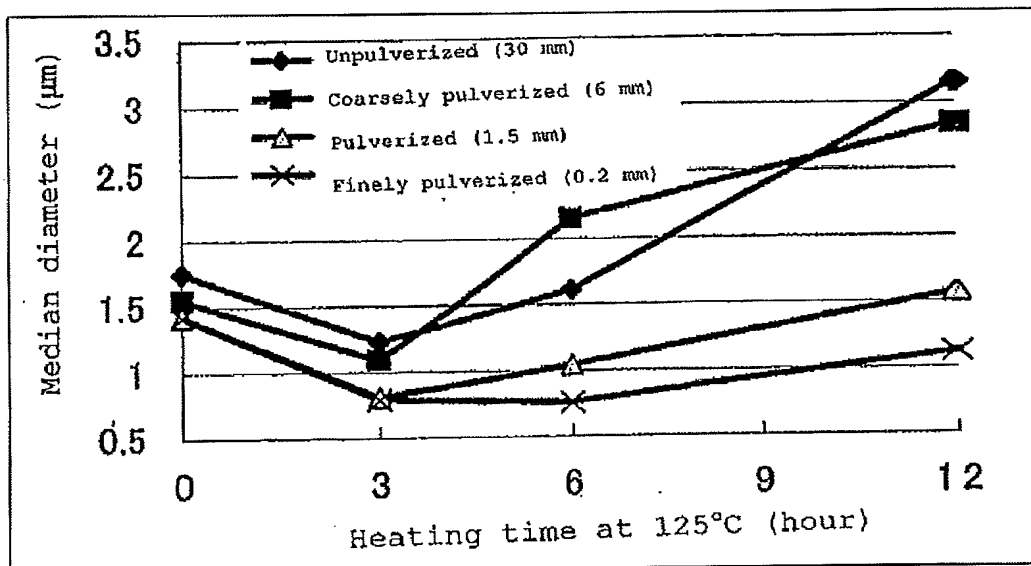


Fig.B

Relationship between particle diameter and emulsifying property under heat at 125°C at reduced pressure



Customer No.
Application No. 11/123.048
Attorney Docket No.:

I hereby declare that all statements made herein of my own knowledge are true and that all statements made on information and belief are believed to be true; and further that these statements were made with the knowledge that willful false statements and the like so made are punishable by fine or imprisonment, or both, under section 1001 of Title 18 of the United States Code, and that such willful false statements may jeopardize the validity of any patent issued on this application.

Date: October 27, 2010

By: Tsuyoshi Katayama
Tsuyoshi KATAYAMA

<Table 1>

Spray-dried powder of gum arabic : average particle diameter 0.064mm

Processing condition : heating at 125 °C for 1 to 8 hours at reduced pressure (about 0.03 atm)

Samples	Form	Processing conditions	Loss-on-drying (%)	Weight average molecular weight (10^4)	AGP (%)	Emulsifying ability Median (μ)	Discoloration	Flavor	Conditions
	Spray-dried powder	Unmodified	11.3	65.0	11.0	1.13	9.3	Little	No change in appearance
1-1	Spray-dried powder	Stirring at 125°C for 1 hour at reduced pressure	0	72.2	12.0	0.78	10.9	Little	No change in appearance
1-2	Spray-dried powder	Stirring at 125°C for 2 hours at reduced pressure	0	87.3	15.1	0.75	13.8	Little	Slight yellowish white but no significant change in appearance
1-3	Spray-dried powder	Stirring at 125°C for 4 hours at reduced pressure	0	108.5	18.5	0.74	16.3	Some	Slight yellowish white but no significant change in appearance
1-4	Spray-dried powder	Stirring at 125°C for 6 hours at reduced pressure	0	144.1	25.2	0.77	18.3	Some	Some yellowish white but no significant change in appearance
1-5	Spray-dried powder	Stirring at 125°C for 8 hours at reduced pressure	0	183.9	30.3	0.77	19.6	Some	Some yellowish white but no significant change in appearance

<Table 2>

Spray-dried powder of gum arabic : average particle diameter 0.064mm

Processing condition : heating at 125 °C for 1 to 8 hours at normal pressure (1 atm)

Samples	Form	Processing conditions	Loss-on-drying (%)	Weight average molecular weight (10 ⁴)	AGP (%)	Emulsifying ability Median (μ)	Discoloration	Flavor	Conditions
	Spray-dried powder	Unmodified	11.3	65.0	11.0	1.13	9.3	Little	No change in appearance
2-1	Spray-dried powder	Stirring at 125°C for 1 hour at normal pressure	10.0	75.5	12.6	0.93	16.0	Some	Browned and caked
2-2	Spray-dried powder	Stirring at 125°C for 2 hours at normal pressure	9.0	92.2	16.6	0.77	19.7	Some	Browned and caked
2-3	Spray-dried powder	Stirring at 125°C for 4 hours at normal pressure	6.6	128.6	23.8	0.77	29.2	Smell of burning	Heavily browned and heavily caked
2-4	Spray-dried powder	Stirring at 125°C for 6 hours at normal pressure	5.8	179.6	30.4	0.87	33.3	Smell of burning	Heavily browned and heavily caked
2-5	Spray-dried powder	Stirring at 125°C for 8 hours at normal pressure	5.5	222.5	37.2	1.12	34.8	Heavy smell of burning	Heavily browned and heavily caked

<Table 3>

Spray-dried powder of gum arabic : average particle diameter 0.064mm

Processing condition :

(1) heating at 90 °C for 0.5 hour at reduced pressure (about 0.03 atm)

(2) heating at 125 °C for 1 to 8 hours at normal pressure (1 atm)

Samples	Form	Processing conditions	Loss-on-drying (%)	Weight average molecular weight (10 ⁴)	AGP (%)	Emulsifying ability Median (μ)	Discoloration	Flavor	Conditions
	Spray-dried powder	Unmodified	11.3	65.0	11.0	1.13	9.3	Little	No change in appearance
	Spray-dried powder	Stirring at 90°C for 0.5 hour at reduced pressure	3.6	65.0	11.0	1.11	9.3	Little	Slightly caked
3-1	Spray-dried powder	Stirring at 125°C for 1 hour at normal pressure	3.2	73.6	12.6	0.89	14.4	Some	Some browned and some caked
3-2	Spray-dried powder	Stirring at 125°C for 2 hours at normal pressure	2.9	92.5	16.0	0.77	16.9	Some	Some browned and some caked
3-3	Spray-dried powder	Stirring at 125°C for 4 hours at normal pressure	2.6	120.3	20.6	0.77	23.0	Some	Browned and caked
3-4	Spray-dried powder	Stirring at 125°C for 6 hours at normal pressure	2.3	162.4	26.9	0.76	26.3	Smell of burning	Heavily browned and heavily caked
3-5	Spray-dried powder	Stirring at 125°C for 8 hours at normal pressure	1.7	200.9	30.9	0.77	28.5	Smell of burning	Heavily browned and heavily caked

<Table 4> → <Table I: DECLARATION UNDER 37 C.F.R. § 1.132>

Processing condition : heating at 125 °C for 3 to 12 hour at reduced pressure (about 0.03 atm)

	Processing conditions	Loss-on-drying (%)	AGP (%)	Weight recovery ratio (%)	Polydispersity (P)	Emulsifying ability Median (μ)	Discoloration	Conditions
Samples 1) Gum arabic (unpulverizates) 30 mm	Unmodified	14.8	14.3	100	2.1	1.75	6.1	No change in appearance
	125°C, 3H	11.1	20.9	96	5.2	1.23	17.1	Dried surface and wet interior
	125°C, 6H	7.0	24.1	93	6.2	1.61	34.8	Dried surface and wet interior
	125°C, 12H	2.7	25.0	80	6.7	3.15	36.6	Dried surface and wet interior
Samples 2) Gum arabic (coarse pulverizates) 6 mm	Unmodified	14.8	15.6	100	2.1	1.55	5.9	No change in appearance
	125°C, 3H	6.5	19.3	98	4.5	1.09	25.9	Dried surface and wet interior
	125°C, 6H	2.8	24.5	96	5.5	2.15	36.5	Dried surface and wet interior
	125°C, 12H	1.3	26.9	93	6.0	2.84	37.5	Dried surface and wet interior
Samples 3) Gum arabic (pulverizates) 1.5 mm	Unmodified	14.5	14.7	100	2.1	1.42	7.0	No change in appearance
	125°C, 3H	2.5	18.2	99	3.8	0.81	12.1	No significant change in appearance
	125°C, 6H	1.4	20.9	98	4.5	1.04	21.1	No change in appearance
	125°C, 12H	0.3	26.5	95	5.3	1.17	25.6	No change in appearance
Samples 4) Gum arabic (Fine pulverizates) 0.083 mm	Unmodified	14.5	14.7	100	2.1	1.42	7.0	No change in appearance
	125°C, 3H	0.5	18.2	100	3.6	0.79	10.5	No change in appearance
	125°C, 6H	0	23.8	99	4.3	0.76	14.6	No change in appearance
	125°C, 12H	0	26.6	97	5.0	1.02	19.5	No change in appearance

<Table 5>

Fine pulverizates of gum arabic : average particle diameter 0.083mm

Processing condition : heating at 125 °C for 1 to 8 hours at reduced pressure (about 0.03 atm)

Sample	Form	Processing conditions	Loss-on-drying (%)	AGP (%)	Emulsifying ability Median (μ)	Discoloration	Polydispersity (P)	Weight recovery ratio (%)
Blank	Fine pulverizates	Unmodified	13.9	14.0	1.03	5.2	2.1	100
(3)-1	Fine pulverizates	Stirring at 125°C for 1 hour at reduced pressure	0.2	15.4	0.78	6.9	2.5	100
(3)-2	Fine pulverizates	Stirring at 125°C for 2 hours at reduced pressure	0	17.0	0.75	9.4	2.9	100
(3)-3	Fine pulverizates	Stirring at 125°C for 4 hours at reduced pressure	0	20.3	0.75	12.2	3.6	100
(3)-4	Fine pulverizates	Stirring at 125°C for 6 hours at reduced pressure	0	25.2	0.81	14.9	4.4	99
(3)-5	Fine pulverizates	Stirring at 125°C for 8 hours at reduced pressure	0	26.6	0.85	17.3	4.8	99

<Table 6>

Spray-dried powder of gum arabic : average particle diameter 0.064mm

Processing condition : heating at 125 °C for 1 to 10 hours at reduced pressure (about 0.03 atm)

Samples	Form	Processing conditions	Loss-on-drying (%)	AGP (%)	Emulsifying ability Median (μ)	Discoloration	Viscosity (mPa•S)	Polydispersity (P)	Weight recovery ratio (%)
Blank	Spray-dried powder	Unmodified	11.3	11.0	1.13	9.3	11.7	2.1	100
(4)-1	Spray-dried powder	Stirring at 125°C for 1 hour at reduced pressure	0	12.0	0.78	10.9	11.3	2.2	100
(4)-2	Spray-dried powder	Stirring at 125°C for 2 hours at reduced pressure	0	15.1	0.75	13.8	12.4	2.5	100
(4)-3	Spray-dried powder	Stirring at 125°C for 4 hours at reduced pressure	0	18.5	0.74	16.3	13.3	3.0	101
(4)-4	Spray-dried powder	Stirring at 125°C for 6 hours at reduced pressure	0	25.2	0.77	18.3	14.3	3.4	100
(4)-5	Spray-dried powder	Stirring at 125°C for 8 hours at reduced pressure	0	30.3	0.77	19.6	16.4	3.8	100
(4)-6	Spray-dried powder	Stirring at 125°C for 10 hours at reduced pressure	0	35.6	0.83	20.9	19.0	4.2	100

<Table 7>

Spray-dried powder of gum arabic : average particle diameter 0.064mm

Processing condition :

(1) heating at 90 °C to 180 °C at reduced pressure (about 0.03 atm)

(2) heating at 70 °C at reduced pressure (about 0.03 atm)

(3) heating at 200°C at reduced pressure (about 0.03 atm)

Samples	Form	Processing conditions	Weight average molecular weight (10 ⁴)	AGP (%)	Emulsifying ability Median (μ)	Discoloration	Flavor	Conditions
Blank	Spray-dried powder	Unmodified	65.0	11.0	1.13	9.3	Little	No change in appearance
Sample 1)	Spray-dried powder	Stirring at 90°C for 24 hours at reduced pressure	98.6	16.8	0.75	13.9	Some	Slightly yellowish white but no change in appearance
Sample 2)	Spray-dried powder	Stirring at 110°C for 24 hours at reduced pressure	105.0	18.9	0.74	15.5	Some	Slightly yellowish white but no change in appearance
Sample 3)	Spray-dried powder	Stirring at 125°C for 4 hours at reduced pressure	108.5	21.7	0.74	16.3	Some	Slightly yellowish white but no change in appearance
Sample 4)	Spray-dried powder	Stirring at 140°C for 2 hours at reduced pressure	132.5	24.5	0.75	17.5	Some	Slightly yellowish white but no change in appearance
Sample 5)	Spray-dried powder	Stirring at 180°C for 10 minutes at reduced pressure	155.2	25.8	0.77	19.8	Smell of burning	Yellowish white but little change in appearance
Sample 6)	Spray-dried powder	Stirring at 70°C for 24 hours at reduced pressure	66.0	11.9	1.12	10.9	No smell of burning	No change in appearance
Sample 7)	Spray-dried powder	Stirring at 200°C for 10 minutes at reduced pressure	187.9	32.5	1.18	42.5	Heavy smell of burning	Heavy yellowish white discoloration, and partially burned black